In addition, new claims 30-44 are added, and these claims correspond to amended claims 1-29 in which the multi-segmented fluoropolymer is limited to a multi-segmented fluoropolymer comprising a block copolymer. Specifically stated, claims 30-37, 38-40, 41 and 42 correspond to claims 1-8, 14-16, 17, and 22, respectively, in which the multi-segmented fluoropolymer is limited to a multi-segmented fluoropolymer comprising a block copolymer. Claim 43 is dependent on claim 29 and contains the feature of claim 15. Claim 44 is dependent on claim 43 and contains the features of claim 16. In claims 11, 17, 18, 22, 26, 41 and 42, the definitions of X, X^1, Y, n and Rf as in claim 4 have now been added as suggested by the Examiner to make these claims clear.

The claims in the present form now overcome the rejection under 35 U.S.C. §112 made by the Examiner. In addition, the claims are also patentable over the prior art for the reasons stated below.

Present Invention

Claim 1 defines a material for a solid polyelectrolyte, comprising a multi-segmented fluoropolymer having:

A fluoropolymer chain segment A containing sulfonic acid functional groups, which is a copolymer comprising:

an ethylenic fluoromonomer unit containing sulfonic acid functional groups represented by a specific formula; and

at least one type of ethylenic fluoromonomer unit copolymerizable with the ethylenic fluoromonomer unit and containing no sulfonic acid functional groups;

and a fluoropolymer chain segment B containing no sulfonic acid functional groups, the fluoropolymer chain segment B having a crystalline melting point of 100°C or higher or a glass transition point of 100°C or higher.

Claim 30 claims a material as defined in present claim 1 in which the multi-segmented fluoropolymer is limited to a multi-segmented fluoropolymer comprising a block copolymer.

In the present invention, the use of the specific copolymer segments A and B, or the block copolymer having the segments A and B improves the mechanical properties, high-temperature mechanical properties, heat resistance, ion conductivity, and other properties of the fluoropolymer. Further, when the fluoropolymer is used as a material for a solid polyelectrolyte in a fuel cell, it effectively improves the heat resistance, durability, and creep resistance, thus increasing the reliability.

Patentability of the Present Invention Over the Prior Art

In the Official Action, the Examiner rejected the claims as anticipated by Masayuki et al. (JP 06-260,184). This rejection, insofar as applied to the claims as amended, is respectfully traversed for the reasons as stated below.

Comparison with Masayuki et al. (JP 06-260,184)

The Masayuki et al. reference discloses the use of a cation exchange membrane comprising a perfluorocarbon copolymer having sulfonic acid groups, in a solid polyelectrolyte fuel cell.

Masayuki et al. mentions a copolymer of $CF_2=CF_2$ and $CF_2=CF-(OCF_2CFX)_m-O_q-(CF_2)_n-A$ as a perfluorocarbon polymer, and employs a copolymer of $CF_2=CF_2$ and $CF_2=CFOCF_2CFCF_3O(CF_2)_2-SO_2F$ in the Examples.

These copolymers are deemed to correspond to the segment A in the present invention. However, Masayuki et al. teaches nothing about a polymer corresponding to the segment B.

Further, Masayuki et al. states, in paragraph [0016], that a polymer was prepared by a method described in JP 02-88,645. In the Examples of JP 02-88,645, $(NH_2)_2S_2O_8$, which is a radical initiator, was used to prepare a polymer, and therefore the polymer of JP 02-88,645 is believed to be a random copolymer. Accordingly, the polymer used in Masayuki et al. is also deemed to be a random copolymer.

In contrast, the fluoropolymer in claim 1 comprises the above-mentioned specific segments A and B, and thus is different from the fluorine-containing random copolymer of Masayuki et al. that merely has a side chain containing sulfonyl groups and/or carboxyl groups. The same applies to claims 2-29, 43 and 44.

Moreover, the fluoropolymer in claim 30 comprises a block copolymer having the segments A and B, and thus differs from the random copolymer of Masayuki et al.. The same applies to claims 31-42.

Masayuki et al. nowhere teaches or suggests the above-mentioned remarkable effects achieved by using the segments A and B or a block copolymer having the segments A and B. Accordingly, the Examiner's rejection on the basis of this reference is respectfully traversed and should be withdrawn.

In the Official Action, the Examiner also rejected the claims as anticipated by the Harada US Patent No. 5,399,184. This rejection, insofar as applied to the claims as amended, is respectfully traversed for the reasons as stated below.

Comparison with Harada (US 5,399,184)

Harada discloses a cation exchange membrane comprising a copolymer represented by formula (1) or (2), which is a copolymer of a tetrafluoroethylene unit and a perfluorovinyl ether unit having sulfonic acid groups on the side chain, for use in a gas diffusion electrode for a solid polyelectrolyte fuel cell.

The copolymer of formula (1) or (2) is a random copolymer, since it is prepared using NAFION SOLUSION sold by Aldrich Chemical Co.

This copolymer is also deemed to correspond to the segment A in the present invention. Harada teaches nothing about a polymer corresponding to the segment B.

In contrast, the fluoropolymer in claim 1 has the specific segments A and B, and thus differs from the random copolymer represented by formula (1) or (2). The same applies to claims 2-29, 43 and 44.

Further, the fluoropolymer in claim 30 comprises a block copolymer having the segments A and B, but Harada does not teach the use of a block copolymer. The same applies to claims 31-42. Harada thus in no way teaches nor suggests that the use of a fluoropolymer having the segments A and B or a fluoropolymer that comprises a block copolymer having the segments A and B achieves the above-mentioned remarkable effects. Accordingly, the Examiner's rejection on the basis of this reference is respectfully traversed and should be withdrawn.

Conclusion

It is thus clear that the inventions as defined in claims 1 and 5-44 are novel and unobvious over the Masayuki et al. and Harada references, and that the Examiner's rejections, insofar as applied to the claims as amended, are respectfully traversed and should be withdrawn.

In view of the foregoing, Applicants respectfully submit that the present application is in condition for immediate allowance, and such action is earnestly solicited.

Respectfully submitted,

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ATTACHMENT A

Marked Up Replacement Claims

Following herewith is a marked up copy of each rewritten claim.

1. (Amended) A material for a solid polyelectrolyte, comprising a multi-segmented fluoropolymer having ~~at least two types of fluoropolymer chain segments differing in monomer composition, at least one type of the fluoropolymer chain segments containing sulfonic acid functional groups~~.

a fluoropolymer chain segment A containing sulfonic acid functional groups, which is a copolymer comprising:

(a) an ethylenic fluoromonomer unit containing sulfonic acid functional groups represented by Formula (1)

$$CX_2=CX^1-(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein X and X^1 may be the same or different and are each hydrogen or fluorine; Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s); and n is 0 or 1; and

(b) at least one type of ethylenic fluoromonomer unit copolymerizable with the unit (a) and containing no sulfonic acid functional groups;

and a fluoropolymer chain segment B containing no sulfonic acid functional groups, the fluoropolymer chain segment B having a crystalline melting point of 100°C or higher or a glass transition point of 100°C or higher

2. Canceled.

3. Canceled.

4. Canceled.

5. (Twice Amended) The material according to claim ~~3~~1, wherein the at least one type of ethylenic fluoromonomer unit (b) containing no sulfonic acid functional groups comprises tetrafluoroethylene.

6. (Amended) The material according to claim ~~2~~1, wherein the fluoropolymer chain segment B is a polymer chain comprising 85 to 100 mol% of tetrafluoroethylene and 15 to 0 mol% of a monomer represented by Formula (3)

$$CF_2=CF\text{-}Rf^a \qquad (3)$$

wherein Rf^a is CF_3 or ORf^b wherein Rf^b is C_1 to C_5 perfluoroalkyl.

7. (Amended) The material according to claim ~~2~~1, wherein the multi-segmented fluoropolymer has an equivalent weight of 400 to 1600.

marked claims

11. (Amended) The material according to claim 10, wherein the ethylenic fluoromonomer unit (c) containing sulfonic acid functional groups is represented by Formula (1)

$$CX_2=CX^1-(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein ~~X, X^1, Y, n and Rf are as defined above~~ X and X^1 may be the same or different and are each hydrogen or fluorine; Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s); and n is 0 or 1.

14. (Twice Amended) A solid polyelectrolyte membrane comprising the multi-segmented fluoropolymer according to claim ~~2~~1.

17. (Amended) A multi-segmented fluoropolymer having a fluoropolymer chain segment A^1 containing sulfonic acid functional groups and a fluoropolymer chain segment B^1 containing no sulfonic acid functional groups, wherein:

the fluoropolymer chain segment A^1 containing sulfonic acid functional groups is a copolymer having a molecular weight of 5000 to 750000 and comprising:

(e) 1 to 50 mol% of at least one type of structural unit represented by Formula (1)

$$CX_2=CX^1-(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein ~~X, X^1, Y, n and Rf are as defined above~~ X and X^1 may be the same or different and are each hydrogen or fluorine; Y is F, Cl and OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s); and n is 0 or 1, and

marked claims

(f) 99 to 50 mol% of at least one type of ethylenic monomer structural unit containing no sulfonic acid functional groups; and

the fluoropolymer chain segment B^1 is a fluoropolymer chain containing at least one type of ethylenic fluoromonomer unit and having a molecular weight of 3000 to 12000000.

18. (Amended) The multi-segmented fluoropolymer according to claim 17, wherein the ethylenic fluoromonomer (e) in the fluoropolymer chain segment A^1 is represented by Formula (2)

$$CF_2=CFO\text{-}Rf\text{-}SO_2Y \qquad (2)$$

wherein ~~Y and Rf are as defined for Formula (1)~~ Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s).

22. (Amended) A multi-segmented fluoropolymer having at least two types of fluoropolymer chain segments C^1 and D^1 containing sulfonic acid functional groups, wherein:

the fluoropolymer chain segment C^1 is a copolymer having a molecular weight of 5000 to 750000 and comprising:

(g) 13 to 50 mol% of at least one type of ethylenic fluoromonomer structural unit containing sulfonic acid functional groups and represented in Formula (1)

$$CX_2=CX^1\text{–}(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein ~~X, X^1, Y, n and Rf are as defined above~~ X and X^1 may be the same or different and are each hydrogen or fluorine; Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal

marked claims

or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s); and n is 0 or 1, and

(h) 87 to 50 mol% of at least one type of ethylenic monomer structural unit containing no sulfonic acid functional groups; and

the fluoropolymer chain segment D^1 is a fluoropolymer chain having a molecular weight of 3000 to 1200000 and comprising:

(i) not less than 0.1 mol% but less than 13 mol% of at least one type of ethylenic fluoromonomer unit containing sulfonic acid functional groups and represented by Formula (1)

$$CX_2=CX^1-(O)_n\text{-Rf-}SO_2Y \qquad (1)$$

wherein X, X^1, Y, n and Rf are as defined above, and

(j) more than 87 mol% but not more than 99.9 mol% of at least one type of ethylenic monomer unit containing no sulfonic acid functional groups.

23. (Amended) The multi-segmented fluoropolymer according to claim 22, wherein the ethylenic fluoromonomer (g) in the fluoropolymer chain segment C^1 is represented by Formula (2)

$$CF_2=CFO\text{-Rf-}SO_2Y \qquad (2)$$

wherein ~~Y and Rf are as defined for Formula (1)~~ Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s).

marked claims

26. (Amended) The multi-segmented fluoropolymer according to claim 22, wherein the ethylenic fluoromonomer (i) in the fluoropolymer chain segment D^1 is represented by Formula (2)

$$CF_2=CFO\text{-}Rf\text{-}SO_2Y \qquad (2)$$

wherein ~~Y and Rf are as defined for Formula (1)~~ Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s).

30. (new) A material for a solid polyelectrolyte, comprising a multi-segmented fluoropolymer that comprises a block copolymer containing at least two types of fluoropolymer chain segments differing in monomer composition, at least one type of the fluoropolymer chain segments containing sulfonic acid functional groups.

31. (new) The material according to claim 30, which comprises a multi-segmented fluoropolymer that comprises a block copolymer containing a fluoropolymer chain segment A containing sulfonic acid functional groups and a fluoropolymer chain segment B containing no sulfonic acid functional groups, the fluoropolymer chain segment B having a crystalline melting point of 100°C or higher or a glass transition point of 100°C or higher.

32. (new) The material according to claim 31, wherein the fluoropolymer chain segment A containing sulfonic acid functional groups is a copolymer comprising:

(a) an ethylenic fluoropolymer unit containing sulfonic acid functional groups; and

(b) at least one type of ethylenic fluoromonomer unit copolymerizable with the unit (a) and containing no sulfonic acid functional groups.

33. (new) The material according to claim 32, wherein the ethylenic fluoromonomer unit (a) containing sulfonic acid functional groups is represented by Formula (1)

$$CX_2=CX^1-(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein X and X^1 may be the same or different and are each hydrogen or fluorine; Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s); and n is 0 or 1.

34. (new) The material according to claim 32, wherein the at least one type of ethylenic fluoromonomer unit (b) containing no sulfonic acid functional groups is tetrafluoroethylene.

35. (new) The material according to claim 31, wherein the fluoropolymer chain segment B is a polymer chain comprising 85 to 100 mol% of tetrafluoroethylene and 15 to 0 mol% of a monomer represented by Formula (3)

$$CF_2=CF\text{-}Rf^a \qquad (3)$$

wherein Rf^a is CF_3 or ORf^b wherein Rf^b is C_1 to C_5 perfluoroalkyl.

36. (new) The material according to claim 31, wherein the multi-segmented fluoropolymer has an equivalent weight of 400 to 1600.

marked claims

37. (new) The material according to claim 8, which comprises a multi-segmented fluoropolymer having a block copolymer of at least two types of fluoropolymer chain segments C and D containing sulfonic acid functional groups, the fluoropolymer chain segment D having a smaller equivalent weight than the fluoropolymer chain segment D.

38. (new) A solid polyelectrolyte membrane comprising the multi-segmented fluoropolymer according to claim 30.

39. (new) The solid polyelectrolyte membrane according to claim 38, wherein the multi-segmented fluoropolymer contains protonated sulfonic acid (SO_3H) groups as the sulfonic acid functional groups, and has a modulus of elasticity of at least $1X10^8$ dyn/cm^2 at 110°C or higher.

40. (new) The solid polyelectrolyte membrane according to claim 39, wherein the equivalent weight of the whole multi-segmented fluoropolymer is 1600 or less.

41. (new) The multi-segmented fluoropolymer according to claim 17, which has a block copolymer of a fluoropolymer chain segment A^1 containing sulfonic acid functional groups and a fluoropolymer chain segment B^1 containing no sulfonic acid functional groups, wherein:

the fluoropolymer chain segment A^1 containing sulfonic acid functional groups is a copolymer having a molecular weight of 5000 to 750000 and comprising:

(e) 1 to 50 mol% of at least one type of structural unit represented by Formula (1)

$$CX_2=CX^1-(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein X and X^1 may be the same or different and are each hydrogen or fluorine; Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_5 alkyl; Rf is C_1 to C_{40} divalent fluoroalkylene or C_1 to C_{40} divalent fluoroalkylene having ether bond(s); and n is 0 or 1, and,

(f) 99 to 50 mol% of at least one type of ethylenic monomer structural unit containing no sulfonic acid functional groups; and

the fluoropolymer chain segment B^1 is a fluoropolymer chain containing at least one type of ethylenic fluoromonomer unit and having a molecular weight of 3000 to 1200000.

42. (new) The multi-segmented fluoropolymer according to claim 22, which has a block copolymer of at least two types of fluoropolymer chain segments C^1 and D^1 containing sulfonic acid functional groups, wherein:

the fluoropolymer chain segment C^1 is a copolymer having a molecular weight of 5000 to 750000 and comprising:

(g) 13 to 50 mol% of at least one type of ethylenic fluoromonomer structural unit containing sulfonic acid functional groups and represented by Formula (1)

$$CX_2=CX^1-(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein X and X^1 may be the same or different and are each hydrogen or fluorine; Y is F, Cl or OY^1 wherein Y^1 is hydrogen, alkali metal or C_1 to C_{40} divalent fluoroalkylene having ether bond(s); and n is 0 or 1, and

marked claims

(h) 87 to 50 mol% of at least one type of ethylenic monomer structural unit containing no sulfonic acid functional groups; and

the fluoropolymer chain segment D^1 is a fluoropolymer chain having a molecular weight of 3000 to 1200000 and comprising:

(i) not less than 0.1 mol% but less than 13 mol% of at least one type of ethylenic fluoromonomer unit containing sulfonic acid functional groups and represented by Formula (a)

$$CX_2=CX^1-(O)_n\text{-}Rf\text{-}SO_2Y \qquad (1)$$

wherein X, X^1, Y, n and Rf are as defined above, and

(j) more than 87 mol% but not more than 99.9 mol% of at least one type of ethylenic monomer unit containing no sulfonic acid functional groups.

43. (new) The solid polyelectrolyte membrane according to claim 29, wherein the multi-segments fluoropolymer contains protonated sulfonic acid (SO_3H) groups as the sulfonic acid functional groups, and has a modulus of elasticity of at least $1X10^8$ dyn/cm^2 at 110°C or higher.

44. (new) The solid polyelectrolyte membrane according to claim 43, wherein the equivalent weight of the whole multi-segmented fluoropolymer is 1600 or less.

marked claims